Analytical Constructions of a Family of Dense Tetrahedron Packings and the Role of Symmetry

S. Torquato[1,2,3,4,5] and Y. Jiao[5]

[1]*Department of Chemistry, Princeton University, Princeton New Jersey 08544, USA*

[2]*Princeton Center for Theoretical Physics, Princeton University, Princeton New Jersey 08544, USA*

[3]*Program in Applied and Computational Mathematics, Princeton University, Princeton New Jersey 08544, USA*

[4]*School of Natural Sciences, Institute for Advanced Study, Princeton NJ 08540 and*

[5]*Department of Mechanical and Aerospace Engineering, Princeton University, Princeton New Jersey 08544, USA*

Abstract

The determination of the densest packings of regular tetrahedra (one of the five Platonic solids) is attracting great attention as evidenced by the rapid pace at which packing records are being broken and the fascinating packing structures that have emerged. We have discovered the densest known packings of regular tetrahedra with a density $\phi = \frac{12250}{14319} = 0.855506\ldots$. These packings are special cases of an analytical two-parameter family of dense periodic packings with four particles per fundamental cell that we have constructed here. From this family, we can recover a set of recent packing arrangements due to Kallus *et al.* with density $\phi = \frac{100}{117} = 0.854700\ldots$, which has higher symmetry than our densest packings. We also describe a procedure that could lead to rigorous upper bounds on the maximal density of tetrahedron packings, which could aid in assessing the packing efficiency of candidate dense packings.

PACS numbers: 61.50.Ah, 05.20.Jj

I. INTRODUCTION

Dense packings of nonoverlapping solid objects or particles are ubiquitous in synthetic and natural situations. Packing problems arise in technological contexts, such as the packaging industries, agriculture (e.g., grains in silos), and solid-rocket propellants, and underlie the structure of a multitude of biological systems (e.g., tissue structure, cell membranes, and phyllotaxis). Dense particle packings are intimately related to the structure of low-temperature states of condensed matter, such as liquids, glasses and crystals [1–4]. In the last decade, scientific attention has broadened from the study of dense packings of spheres (the simplest shape that does not tile Euclidean space) [5–14] to dense packings of nonspherical particles [15–17]. A basic characteristic of a packing in d-dimensional Euclidean space $\mathbb{R}^d$ is its density ϕ, defined to be the fraction of $\mathbb{R}^d$ that is covered by the particles.

A problem that has been of great scientific interest for centuries is the determination of the densest arrangement(s) of particles that do not tile space and the associated maximal density ϕ_{max}. For generally shaped particles, finding the densest packings is notoriously difficult. This salient point is summarized well by Henry Cohn [18] who recently remarked, "For most grain shapes we cannot guess or even closely approximate the answer, let alone prove it, and it is difficult to develop even a qualitative understanding of the effects of grain shape on packing density." Until recently, very little was known about the densest packings of polyhedral particles. The difficulty in obtaining dense packings of polyhedra is related to their complex rotational degrees of freedom and to the non-smooth nature of their shapes [19, 20].

Recently, we set out to attempt to determine the densest known packings of the Platonic and Archimedean solids [19, 20]. It was shown that the central symmetry of the majority of the Platonic and Archimedean solids distinguish their dense packing arrangements from those of the non-centrally symmetric ones in a fundamental way. (A particle is centrally symmetric if it has a center C that bisects every chord through C connecting any two boundary points of the particle; i.e., the center is a point of inversion symmetry.) The tetrahedron is the only Platonic solid that lacks central symmetry, an attribute that geometrically frustrates it to a greater degree than the majority of the remaining solids in this set that do not tile space [20]. A number of organizing principles emerged in the form of conjectures for polyhedra as well as other nonspherical shapes. In the case of polyhedra, the following two

are particularly relevant:

- **Conjecture 1**: *The densest packings of the centrally symmetric Platonic and Archimedean solids are given by their corresponding optimal Bravais lattice packings.*
- **Conjecture 2**: *The densest packing of any convex, congruent polyhedron without central symmetry generally is not a (Bravais) lattice packing, i.e., set of such polyhedra whose optimal packing is not a Bravais lattice is overwhelmingly larger than the set whose optimal packing is a Bravais lattice.*

Conjecture 1 is the analog of Kepler's sphere conjecture for the centrally symmetric Platonic and Archimedean solids. In this sense, such solids behave similarly to spheres in that their densest packings are lattice arrangements and (except for the cube) are geometrically frustrated like spheres. Conjecture 2 has been shown by Conway and Torquato [21] to be true for both the tetrahedron and Archimedean non-centrally symmetric truncated tetrahedron, the latter of which can be arranged in a "uniform" non-Bravais lattice packing with density at least as high as $23/24 = 0.958333\ldots$. (A uniform packing, defined more precisely below, has a symmetry that takes one tetrahedron to another.) Indeed, it was this investigation that has spurred the flurry of activity in the last several years to find the densest packings of tetrahedra. There have been many twists and unexpected turns since 2006 that have led to the densest known packings of tetrahedra that we report here with $\phi = \frac{12250}{14319} = 0.855506\ldots$, which is the highest density obtained from our two-parameter family of constructions described below. Therefore, to place our present results in their proper context, it is instructive to review briefly the developments since 2006.

First, we note that the densest Bravais-lattice packing of tetrahedra (requiring one tetrahedron per fundamental cell such that each tetrahedron in the packing has the same orientation as the others) has a packing fraction $\phi = 18/49 = 0.367\ldots$ and each tetrahedron touches 14 others [25]. Conway and Torquato [21] showed that the densest packings of tetrahedra cannot be Bravais lattices by analytically constructing several such packings with densities that are substantially larger than 0.367. (A non-Bravais lattice packing contains multiple particles, with generally different orientations, per fundamental cell, which is periodically replicated in $\mathbb{R}^d$.) One such packing is a "uniform" packing with density $\phi = 2/3$ and two particles per fundamental cell. The so-called "Welsh" packing has a density $\phi = 0.708333\ldots$

TABLE I: A brief summary of the dense non-lattice packings of tetrahedra. The name of the packing is given along with the year that it was discovered. Here ϕ is the packing density and N is the number of tetrahedra per fundamental cell.

Packing	Year	ϕ	N
Uniform I [21]	2006	$\frac{2}{3} = 0.666666\ldots$	2
Welsh [21]	2006	$\frac{17}{24} = 0.708333\ldots$	34
Icosahedral [21]	2006	$0.716559\ldots$	20
Uniform II [22]	2009	$\frac{139+40\sqrt{10}}{369} = 0.719488\ldots$	2
Wagon Wheels [23]	2008	$0.778615\ldots$	18
Improved Wagon Wheels [19]	2009	$0.782021\ldots$	72
Disordered Wagon Wheels [20]	2009	$0.822637\ldots$	314
Ring Stacks [24]	2009	$0.8503\ldots$	82
Uniform III [22]	2009	$\frac{100}{117} = 0.854700\ldots$	4
Dimer-Uniform	2009	$\frac{12250}{14319} = 0.855506\ldots$	4

and 34 particles per fundamental cell. Yet another non-Bravais lattice packing with density $\phi = 0.716559\ldots$ is based on the filling of "imaginary" icosahedra with the densest arrangement of 20 tetrahedra and then arranging the imaginary icosahedra in their densest lattice packing configuration. The densities of both the Welsh and Icosahedral packings can be further improved by certain particle displacements [21]. Using imperfect "tetrahedral" dice, Chaikin et al. [26] experimentally generated jammed disordered packings with $\phi \approx 0.75$. Employing physical models and a computer algebra system, Chen [23] discovered a remarkably dense periodic arrangement of tetrahedra with $\phi = 0.7786\ldots$, which exceeds the density ($\phi_{max} = \pi/\sqrt{18} = 0.7404\ldots$) of the densest sphere packing by an appreciable amount. We have called this the "wagon-wheels" packing [19, 20].

Torquato and Jiao [19] devised and applied an optimization scheme, called the adaptive-shrinking-cell (ASC) method, that used an initial configuration based on the wagon-wheels packing to yield a non-Bravais lattice packing consisting of 72 tetrahedra per fundamental cell with a density $\phi = 0.782\ldots$ [19]. Using 314 particles per fundamental cell and starting from an "equilibrated" low-density liquid configuration, the same authors were able to improve the density to $\phi = 0.823\ldots$ [20]. This packing arrangement interestingly lacks long-

range order. Haji-Akbari *et al.* [24] numerically constructed a periodic packing of tetrahedra made of parallel stacks of "rings" around "pentagonal" dipyramids consisting of 82 particles per fundamental cell and a density $\phi = 0.8503\ldots$. More recently, Kallus *et al.* [22] found a remarkably simple uniform packing of tetrahedra with high symmetry consisting of only four particles per fundamental cell with density $\phi = \frac{100}{117} = 0.854700\ldots$. Table I summarizes some of the packing characteristics of the non-Bravais lattice packings of tetrahedra.

II. TWO-PARAMETER FAMILY OF DENSE PACKINGS OF TETRAHEDRA

Inspired by the work of Kallus et al. [22], we have applied the adaptive-shrinking-cell (ASC) optimization scheme to examine comprehensively packings with a considerably small number of particles per fundamental cell (from 2 to 32) than we have used in the past. The ASC scheme employs both a sequential search of the configurational space of the particles and the space of lattices via an adaptive fundamental cell that deforms and shrinks on average to obtain dense packings A dense packing with 8-particle basis that emerged from this numerical investigation suggested that it was composed of two very similar fundamental cells, each containing 4 particles. Using one of the 4-particle basis configurations, we were able to find packings with density $\phi = 0.8551034\ldots$ that exceeded the highest density packings with $\phi = 100/117 = 0.854708\ldots$ constructed by Kallus *et al.* Even though our packings possess a type of point inversion symmetry, they are not as symmetric as the densest packings reported in Ref. [22], as we now explain. The four tetrahedra in the fundamental cell in our dense numerically generated packings formed two contacting "dimers". A dimer is composed of a pair of regular tetrahedra with unit edge length that exactly share a common face. The compound object consisting of the two contacting dimers possesses point inversion symmetry, with the inversion center at the centroid of the contacting region on the faces. A Bravais lattice possesses point inversion symmetry about the lattice points and the centroids of the fundamental cells. By placing the symmetry center of the two-dimer compound on the centroids (or the lattice points), we construct packings that generally possess point inversion symmetry only about the symmetry centers of the two-dimer compound. We call such structures *dimer-uniform* packings, since the inversion symmetry acts to take any *dimer* into another. Such packings should be distinguished from the more symmetric *uniform* (or transitive) packings of tetrahedra in which the symmetry operation acts to take any

tetrahedron into another, such as the ones found in Refs. [21] and [22] (see Table I). The latter have almost as much symmetry as a Bravais lattice, except that the centroids of the particles are not just characterized by simple translational symmetry.

We then set out to obtain analytical constructions based on our numerical packings that relax the symmetry conditions on the contacting dimers. In particular, we orient the 3-fold rotational symmetry axis of one of the dimers in an arbitrary direction (say the z-direction of a Cartesian coordinate system), and then fix the origin of the lattice vectors at the centroid of this dimer. (The centroid is located at the center of the contacting faces of the two tetrahedra that comprise the dimer.) Then we place the second dimer in contact with the first one such that there is a center of inversion symmetry that takes one dimer to the other, which implies face-to-face contacts between the two dimers.

The problem of determining the analytical constructions then amounts to determining 12 equations for the 12 unknowns. Nine of the 12 unknowns arise from the three unknown lattice vectors, each of which contains three unknown components. The other 3 unknowns derive from the components of the centroid of the second dimer.

In particular, we let the centroid of the dimer at the origin denoted by $\mathbf{v}_o$, and the centroid of the other dimer denoted by $\mathbf{v}_c$. The vertices of the two dimers at $\mathbf{v}_o$ and $\mathbf{v}_c$ are given by $\mathbf{r}_A = (\frac{1}{2}, \frac{1}{2\sqrt{3}}, 0)$, $\mathbf{r}_B = (-\frac{1}{2}, \frac{1}{2\sqrt{3}}, 0)$, $\mathbf{r}_C = (0, -\frac{1}{\sqrt{3}}, 0)$, $\mathbf{r}_D = (0, 0, \sqrt{\frac{2}{3}})$, $\mathbf{r}_E = (0, 0, -\sqrt{\frac{2}{3}})$ and $\mathbf{r}_A^* = -\mathbf{r}_A + \mathbf{v}_c$, $\mathbf{r}_B^* = -\mathbf{r}_B + \mathbf{v}_c$, $\mathbf{r}_C^* = -\mathbf{r}_C + \mathbf{v}_c$, $\mathbf{r}_D^* = -\mathbf{r}_D + \mathbf{v}_c$, $\mathbf{r}_E^* = -\mathbf{r}_E + \mathbf{v}_c$, respectively. In addition, let the lattice vectors be $\mathbf{v}_1$, $\mathbf{v}_2$, and $\mathbf{v}_3$. The 12 components of the four vectors ($\mathbf{v}_i$, i=c,1,2,3) are the aforementioned unknowns that are related to each other through the nonoverlapping conditions.

In our packings, each dimer has 8 face-to-face contacts and *at least* 2 edge-to-edge contacts. Among these 20 contacts of the two dimers in the fundamental cell, there are 8 independent face-to-face contacts and 1 independent edge-to-edge contact, which reduces the number of variables for the packing from 12 to 3.

A face-to-face contact requires that the projection of the vector distance between the centroids of the two dimers on the contacting face normal is a constant. The 8 independent face-to-face contacts are between the dimer pairs with the centroids at $\{\mathbf{v}_c, \mathbf{v}_o\}$, $\{\mathbf{v}_c, \mathbf{v}_o+\mathbf{v}_1\}$, $\{\mathbf{v}_c, \mathbf{v}_o + \mathbf{v}_2\}$, $\{\mathbf{v}_o + \mathbf{v}_3, \mathbf{v}_c\}$, $\{\mathbf{v}_o + \mathbf{v}_3, \mathbf{v}_c - \mathbf{v}_2\}$, $\{\mathbf{v}_o + \mathbf{v}_3, \mathbf{v}_c + \mathbf{v}_1 - \mathbf{v}_2\}$, $\{\mathbf{v}_o, \mathbf{v}_c - \mathbf{v}_1 + \mathbf{v}_3\}$,

and $\{\mathbf{v}_o, \mathbf{v}_c + \mathbf{v}_1 - \mathbf{v}_2 - 2\mathbf{v}_3\}$. The contact between dimer pairs at $\{\mathbf{v}_i, \mathbf{v}_j\}$ requires

$$(\mathbf{v}_i - \mathbf{v}_j) \cdot \mathbf{n}_{ij} = \frac{2\sqrt{6}}{9}, \tag{1}$$

where $\mathbf{n}_{ij}$ is unit outward contacting face normal of the dimer at $\mathbf{v}_j$.

The edge-to-edge contact requires that the projection of the vector connecting the corresponding ends of two edges on the common perpendicular line of the two edges equals zero, i.e.,

$$[\mathbf{r}_A - (\mathbf{r}_B - \mathbf{v}_1 + \mathbf{v}_2 + \mathbf{v}_3)] \cdot \mathbf{l}_0 = 0, \tag{2}$$

where $\mathbf{l}_0 = (\mathbf{r}_A - \mathbf{r}_D) \times (\mathbf{r}_B - \mathbf{r}_D)$.

Moreover, there are two independent nonoverlapping conditions obtained from 4 *possible* edge-to-edge contacts between neighboring particles, i.e.,

$$[\mathbf{r}_C - (\mathbf{r}_B - \mathbf{v}_1 + \mathbf{v}_3)] \cdot \mathbf{l}_1 \geq 0, \tag{3}$$

$$[\mathbf{r}_C - (\mathbf{r}_D + \mathbf{v}_3)] \cdot \mathbf{l}_2 \geq 0, \tag{4}$$

where $\mathbf{l}_1 = (\mathbf{r}_B - \mathbf{r}_D) \times (\mathbf{r}_E - \mathbf{r}_C)$ and $\mathbf{l}_2 = (\mathbf{r}_D - \mathbf{r}_A) \times (\mathbf{r}_C - \mathbf{r}_E)$.

Furthermore, there are two additional nonoverlapping conditions given by 4 potential vertex-to-face contacts, i.e.,

$$\mathbf{v}_2 \cdot \mathbf{n}_1 = \sqrt{\frac{2}{3}}, \tag{5}$$

where $\mathbf{n}_1 = (\frac{\sqrt{6}}{3}, -\frac{2\sqrt{2}}{6}, -\frac{1}{3})$ is unit outward normal of the contacting face,

$$(\mathbf{v}_1 - \mathbf{v}_2) \cdot \mathbf{n}_2 = \sqrt{\frac{2}{3}}, \tag{6}$$

and $\mathbf{n}_2 = (-\frac{\sqrt{6}}{3}, -\frac{2\sqrt{2}}{6}, \frac{1}{3})$ is unit outward normal of the contacting face.

The edge-to-edge and vertex-to-face contacts are realized when the equality holds in the above conditions (2)-(6). However, these contacts can not be realized simultaneously in general. In particular, Eqs. (1), (2), (3), (5) and Eqs. (1), (2), (4), (6) provide two sets of equations that lead to a family of dense tetrahedral packings whose structures are determined by two parameters a and b, with the corresponding density dependent only on the parameter a. In other words, each density is associated with a spectrum of different packing structures.

Finally, we arrive at an explicit expression for the packing density:

$$\phi = \frac{V_T}{|\mathbf{v}_1 \times \mathbf{v}_2 \cdot \mathbf{v}_3|}, \tag{7}$$

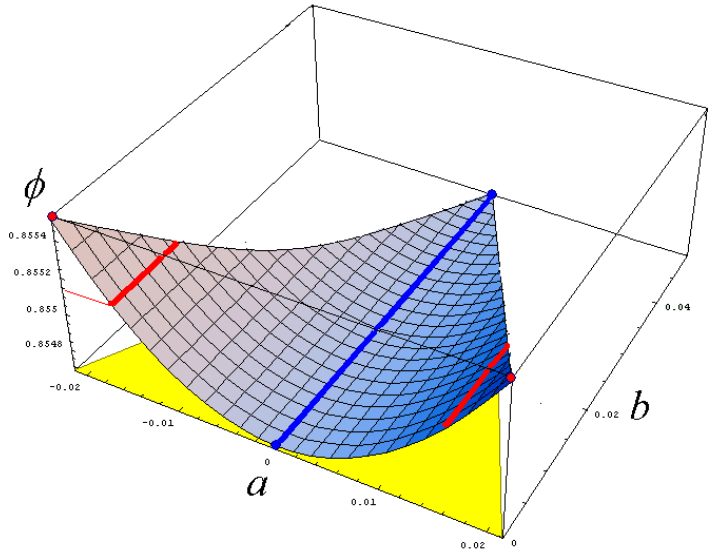


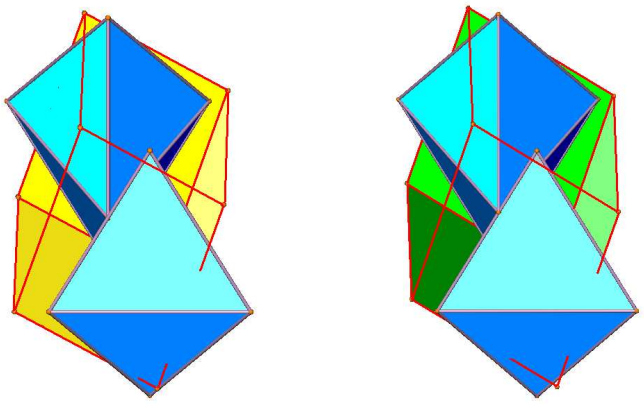
FIG. 1: (color online). The density ϕ surface of our family of tetrahedral packings as a function of the two parameters a and b. As explained in the text, the thick red lines show two sets of tetrahedral packings with distinct structures but with the same density. The two red points correspond to the densest known two tetrahedral packings. The packings found by Kallus *et al.* [22] are recovered from our two-parameter family (thick blue line).

where $V_T = \sqrt{2}/12$ is the volume of a regular tetrahedron with unit edge length. Substituting the lattice vectors expressed in terms of the two parameters a and b into (7) yields

$$\phi = \frac{100}{117 - 240a^2}, \tag{8}$$

where $a \in (-\frac{3}{140}, \frac{3}{140})$. It is important to note that for each $a \neq 0$, there are two sets of packings of tetrahedra, each with distinct structures but possessing the same density (as shown in Fig. 1 by the thick red lines). For $-\frac{3}{140} < a < 0$, the packings are specified by

$$\begin{aligned}
\mathbf{v}_c &= (\frac{1}{5} + \frac{a}{3}, -\frac{4}{5\sqrt{3}}, -\frac{3\sqrt{2}}{5\sqrt{3}} + \sqrt{\frac{2}{3}}a), \\
\mathbf{v}_1 &= (-a, -\frac{\sqrt{3}}{2}, \sqrt{\frac{2}{3}}a), \\
\mathbf{v}_2 &= (\frac{3}{4} + a - b, -\frac{\sqrt{3}}{4} + \sqrt{3}a, -\frac{3}{5\sqrt{6}} - \frac{8}{\sqrt{6}}a + \sqrt{6}b), \\
\mathbf{v}_3 &= (-\frac{7}{20} - 2a + b, -\frac{\sqrt{3}}{4} - \sqrt{3}a, -\frac{9}{5\sqrt{6}} + \frac{10}{\sqrt{6}}a - \sqrt{6}b),
\end{aligned} \tag{9}$$



(a) (b)

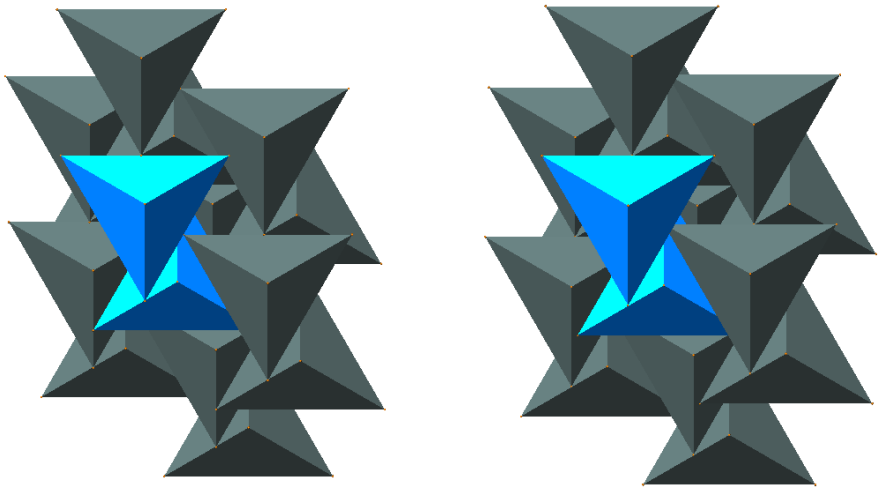
FIG. 2: (color online). Two different configurations of the densest known packings of four tetrahedra (two dimers) within their corresponding rhombohedral fundamental cells: (a) Here $a = -3/140$ and $b = 0$, and the fundamental cell is colored yellow and its boundaries are colored red. (b) Here $a = 3/140$ and $b = 0$, and fundamental cell is colored blue and its boundaries are colored red. The two packings shown in (a) and (b) are only slightly different from one another. Specifically, the difference between the coordinates of the centroids that are not at the origin is $(1/70, 0, \sqrt{6}/70)$. The differences between the shapes of the rhombohedral fundamental cells in the two cases are readily apparent.

where $0 < b < \frac{3+140a}{60}$. For $0 < a < \frac{3}{140}$, the packings are specified by

$$\begin{aligned}
\mathbf{v}_c &= (\frac{1}{5} + \frac{a}{3}, -\frac{4}{5\sqrt{3}}, -\frac{3\sqrt{2}}{5\sqrt{3}} + \sqrt{\frac{2}{3}}a), \\
\mathbf{v}_1 &= (-a, -\frac{\sqrt{3}}{2}, \sqrt{\frac{2}{3}}a), \\
\mathbf{v}_2 &= (\frac{3}{4} - 2a - b, -\frac{\sqrt{3}}{4} + \sqrt{3}a, -\frac{3}{5\sqrt{6}} + \frac{10}{\sqrt{6}}a + \sqrt{6}b), \\
\mathbf{v}_3 &= (-\frac{7}{20} + a + b, -\frac{\sqrt{3}}{4} - \sqrt{3}a, -\frac{9}{5\sqrt{6}} - \frac{8}{\sqrt{6}}a - \sqrt{6}b),
\end{aligned} \tag{10}$$

where $0 < b < \frac{3-140a}{60}$.

The densest packings are associated with $a = -\frac{3}{140}$, $b = 0$ and $a = \frac{3}{140}$, $b = 0$, possessing a density $\phi_{max} = \frac{12250}{14319} = 0.855506....$ In each set, there is a unique packing structure associated with ϕ_{max} (shown as the red points in Fig. 1), instead of a spectrum of structures. Two different configurations of the densest known packings of four tetrahedra within their corresponding rhombohedral fundamental cells are shown in Fug. 2. Figure 3 depicts peri-



(a) (b)

FIG. 3: (color online). This figure shows periodic replicates the densest known tetrahedral packings corresponding to Fig. 2 with 8 fundamental cells (2 along each lattice vector. The four tetrahedra within a fundamental cell are shown in blue: (a) $a = -3/140$ and $b = 0$. (b) $a = 3/140$ and $b = 0$. Note that in (a), the dimer with centroid at $\mathbf{v}_1$ is slightly shifted to the right with respect to the dimer at the origin; and in (b), the dimer with the centroid at $\mathbf{v}_1$ is slightly shifted to the left with respect to the dimer at the origin.

odic replicates of our optimal tetrahedral packings corresponding to those shown in Fig. 2 with 8 fundamental cells (2 along each lattice vector).

At $a = 0$, there is only one set of packings with the same $\phi = 100/117 = 0.85470...$, whose structures are dependent on b (shown as the blue line in Fig. 1). These packings reduce exactly to those discovered by Kallus et al., which possess relatively high symmetry. In particular, $a = 0$ allows the centroids of the dimers, which are related to each other by an integer multiple of $\mathbf{v}_1$, to be perfectly aligned on two of the mirror image planes of the dimers simultaneously, which leads to additional two-fold rotational symmetry of the packing. This additional rotational symmetry, together with the point inversion symmetry, leads to uniform packings with respect to each tetrahedron (not just each dimer), i.e., the symmetry operation acts to take each tetrahedron into another.

III. TOWARDS UPPER BOUNDS ON THE MAXIMAL DENSITY

The problem of determining upper bounds on the maximal density of packings of nonspherical particles is highly nontrivial, and yet such estimates would be indispensable in assessing the packing efficiency of a candidate dense packing, especially if tight upper bounds could be constructed. It has recently been shown that ϕ_{max} of a packing of congruent nonspherical particles of volume v_P in $\mathbb{R}^3$ is bounded from above according to

$$\phi_{max} \leq \min\left[\frac{v_P}{v_S}\,\frac{\pi}{\sqrt{18}}, 1\right], \tag{11}$$

where v_S is the volume of the largest sphere that can be inscribed in the nonspherical particle and $\pi/\sqrt{18}$ is the maximal sphere-packing density [19, 20]. The upper bound (11) will be relatively tight for packings of nonspherical particles provided that the *asphericity* γ (equal to the ratio of the circumradius to the inradius) of the particle is not large. However, for tetrahedra, the asphericity is too large for the upper bound (11) to yield a result that is less than unity.

One possible approach to obtaining nontrivial upper bounds is to attempt to generalize the idea that Rogers used to prove upper bounds on ϕ_{max} for sphere packings [27]. The key concept is to consider a locally dense cluster of 4 contacting spheres in a tetrahedral arrangement and then prove that the fraction of space covered by the spheres within the tetrahedron joining the sphere centers is an upper bound on ϕ_{max}. This can be done because one can triangulate any sphere packing to decompose it into generally irregular tetrahedra with vertices at sphere centers. The fact that the regular tetrahedron has the best density for any tetrahedron, then yields an upper bound for the density of any sphere packing. In the case of the non-tiling Platonic and Archimedean solids, a natural choice for the enclosing region associated with the cluster is its convex hull.

For tetrahedra, we must identify the *least* densest local cluster with density that exceeds ϕ_{max}. A trivial choice is a dimer because the fraction of space covered by the dimer within its convex hull is unity. A nontrivial choice is a 5-particle "wagon-wheel" cluster as shown in Fig. 4. The convex hull of the local packing of five tetrahedra sharing a common edge (a "wagon wheel" cluster) can be decomposed into five regular tetrahedra and five thin irregular tetrahedra. We assume the gaps between the regular tetrahedra are equal, i.e, the thin irregular tetrahedra are congruent. Note the regular tetrahedron shares two faces with

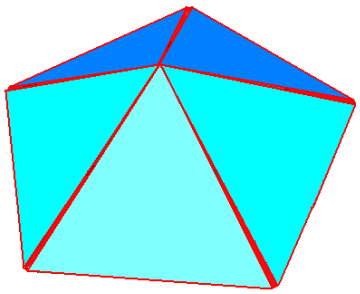


FIG. 4: (color online). The convex hull of five regular tetrahedra in a "wagon wheel" arrangement. The convex hull can be decomposed into five regular tetrahedra (shown in blue) and five thin irregular tetrahedra (shown in red).

its neighboring irregular tetrahedra. Thus, the volume ratio is equal to the ratio of the corresponding heights of the regular and irregular tetrahedron, i.e.,

$$\gamma = \frac{V_T}{V_{T_*}} = \frac{1}{\frac{\sqrt{2}}{2}(3\cos^2\frac{3\pi}{10} - 1)}, \tag{12}$$

where V_T and V_{T_*} is the volume of the regular and irregular tetrahedron, respectively. Thus, the density of this local packing, defined as the fraction space covered by the regular tetrahedra within the convex hull is given by

$$\phi_W = \frac{V_T}{V_T + V_{T_*}} = \frac{1}{\frac{3\sqrt{2}}{2}\cos^2\frac{3\pi}{10} + (1 - \frac{\sqrt{2}}{2})} = 0.974857\ldots. \tag{13}$$

Because this cluster is highly anisotropic and is an effectively "flat" object, it is reasonable to assume that it is not the least densest local cluster and therefore its local density is a gross overestimate of ϕ_{max}.

Since tetrahedra are fully three-dimensional objects, it seems reasonable to assume that the least densest local cluster should be more "isotropic" than the highly anisotropic and effectively "two-dimensional" wagon-wheel cluster. One plausible choice for such a cluster consists of 20 tetrahedra sharing a common vertex (i.e., an icosahedral-like cluster). The convex hull of this cluster can be decomposed into 20 regular tetrahedra, 12 pyramids with pentagonal bases and 30 pyramid with rectangular bases (see Fig. 5). We assume the gaps between tetrahedra are equal and thus the two types of pyramids are congruent. The volume

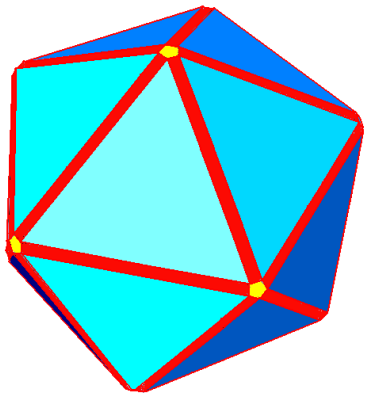


FIG. 5: (color online). The convex hull of 20 regular tetrahedra in an "icosahedral" arrangement. The convex hull can be decomposed into 20 regular tetrahedra (shown in blue), 12 pyramids with pentagonal bases (shown in yellow) and 30 pyramid with rectangular bases (shown in red).

of the regular tetrahedron is $V_T = \frac{\sqrt{2}}{12}$ and the volume V_P of the pyramid with pentagonal base is given by

$$V_P = \frac{5}{12}\tan(\frac{3\pi}{10})L^2\sqrt{1-\frac{L^2}{4\cos^2\frac{3\pi}{10}}}, \tag{14}$$

and the volume V_R of the pyramid with rectangular base is given by

$$V_R = \frac{\sqrt{2}}{6}L\sqrt{1-L^2}, \tag{15}$$

where

$$L = (\frac{2\sqrt{2}}{\tau^2}-1)\sqrt{\frac{1}{6}+\frac{\sqrt{5}}{18}}, \tag{16}$$

and $\tau = (1+\sqrt{5})/2$ is the golden ratio. Thus, the local packing density is given by

$$\phi_I = \frac{20V_T}{20V_T+12V_P+30V_R} = 0.880755\ldots. \tag{17}$$

Note in the above calculations, we have assumed that the gaps between the tetrahedra are equal, which is sufficient to provide an estimate of the fraction of space covered by the cluster within its convex hull. However, the idea of Rogers to prove an upper bound for sphere packings cannot be used here because there is no analogous decomposition of space into irregular convex hulls shown in Figs. 4 or 5. A completely new idea is needed to prove

that the aforementioned estimates are bounds, but it seems plausible that they are correct bounds. If one could prove that these clusters are indeed locally denser than the globally densest packings, then the aforementioned estimates provide upper bounds on ϕ_{max}, but they cannot be sharp, i.e., they are not achievable by any packings. It is noteworthy the density $\phi = 0.855506\ldots$ of our our densest packings is relatively close to this putative upper bound density estimate of $0.880755\ldots$.

IV. DISCUSSION

For all of the small systems (including 2 to 32 particles) that we investigated using our numerical ASC scheme, the densest packings that emerged had a 4-particle basis. Our analytical constructions indicate that for such small systems, the highest density packings we found could be optimal. However, there is no reason to believe that denser packings could not be discovered by carrying out exhaustive searches to determine the globally maximal densities of packings with successively larger numbers of particles per fundamental cell. Previous numerical studies have indicated that dense packings may have a large number of particles per fundamental cell arranged in a complex fashion, e.g., the "disordered wagon-wheels" packing with $\phi = 0.822637\ldots$ [20] and the "ring stacks" packing with $\phi = 0.8503\ldots$ [24]. However, it is plausible that such packings are in fact only locally optimal solutions and hence the numerical techniques used to obtain them are incapable of extricating themselves from these "trapped" regions of configuration space to find denser and more ordered structures due to the intrinsic geometrical frustration of the tetrahedron. This may also call into question claims made by Haji-Akbari *et al.* [24] that their packings, which are characterized by an effective "quasicrystal-like" plane, are true thermodynamic equilibrium phases of tetrahedra, especially at high densities. Instead, our highest-density constructions suggest that uniform periodic packings with a 4-particle basis (or even some yet unknown denser periodic packing) and unjammmed, lower-density countyerparts could be the stable phases at such high densities. If the latter is correct and the putative "quasicrystal-like" phase truly exists at intermediate densities, then it is hard to imagine how a quasicrystal-like structure of tetrahedra under quasi-static compression (densification) could rearrange to a structure with a more ordered periodic arrangement with higher symmetry. However, it is difficult to draw such definitive conclusions without further study.

Although there could still be tetrahedral packings denser than our constructions, it appears that all of the evidence thus far points to the fact that the densest tetrahedral packings cannot possess very high symmetry [19–21, 23, 24] due to the lack of central symmetry of a tetrahedron and because tetrahedra cannot tile space [20]. Indeed, our densest uniform 4-particle-basis packings found in the present paper improved upon the best analogous packings of Kallus *et al.* [22] by relaxing the two-fold rotational symmetry constraints they imposed.

Acknowledgments

We are grateful to Henry Cohn, Yoav Kallus and John Conway for helpful discussions. S. T. thanks the Institute for Advanced Study for its hospitality during his stay there. This work was supported by the Division of Mathematical Sciences at the National Science Foundation under Award Number DMS-0804431 and by the MRSEC Program of the National Science Foundation under Award Number DMR-0820341.

---

[1] J. D. Weeks, D. Chandler, and H. C. Andersen, J. Chem. Phys. **54**, 5237 (1971).

[2] R. Zallen, *The Physics of Amorphous Solids* (Wiley, New York, 1983).

[3] P. M. Chaikin and T. C. Lubensky, *Principles of Condensed Matter Physics* (Cambridge University Press, New York, 1995).

[4] S. Torquato, *Random Heterogeneous Materials: Microstructure and Macroscopic Properties* (Springer-Verlag, New York, 2002).

[5] C. F. Gauss, Göttingsche Gelehrte Anzeigen (1831), see also J. reine angew. Math., vol. 20, 1840, 312-320.

[6] J. D. Bernal, Nature **185**, 68 (1960).

[7] J. H. Conway and N. J. A. Sloane, *Sphere Packings, Lattices and Groups* (Springer-Verlag, New York, 1998).

[8] A. J. Liu and S. R. Nagel, Nature **396**, 21 (1998).

[9] S. Torquato, T. M. Truskett, and P. G. Debenedetti, Phys. Rev. Lett. **84**, 2064 (2000).

[10] C. S. O'Hern, S. A. Langer, A. J. Liu, and S. R. Nagel, Phys. Rev. Lett. **88**, 075507 (2002).

[11] H. Cohn and N. Elkies, Annals Math. **157**, 689 (2003).

[12] T. C. Hales, Ann. Math. **162**, 1065 (2005).

[13] S. Torquato and F. H. Stillinger, Experimental Math. **15**, 307 (2006).

[14] H. Cohn and A. Kumar, Ann. Math. **170**, 1003 (2009).

[15] U. Betke and M. Henk, Comput. Geom. **16**, 157 (2000).

[16] A. Donev, F. H. Stillinger, P. M. Chaikin, and S. Torquato, Phys. Rev. Lett. **92**, 255506 (2004).

[17] Y. Jiao, F. H. Stillinger, and S. Torquato, Phys. Rev. E **79**, 041309 (2009).

[18] H. Cohn, Nature **460**, 801 (2009).

[19] S. Torquato and Y. Jiao, Nature **460**, 876 (2009).

[20] S. Torquato and Y. Jiao, Phys. Rev. E **80**, 041104 (2009).

[21] J. H. Conway and S. Torquato, Proc. Nat. Acad. Soc. **103**, 10612 (2006).

[22] Y. Kallus, V. Elser, and S. Gravel (2009), arXiv:0910.5226.

[23] E. R. Chen, Discrete Comput. Geom. **40**, 214 (2008).

[24] A. Haji-Akbari, M. Engel, A. S. Keys, X. Zheng, R. G. Petschek, P. Palffy-Muhoray, and S. C. Glotzer, Nature **462**, 773 (2009).

[25] D. J. Hoylman, Bull. Amer. Math. Soc. **76**, 135 (1970).

[26] P. M. Chaikin, S. Wang, and A. Jaoshvili, in *Am. Phys. Soc. March Meeting* (2007).

[27] C. A. Rogers, Proc. Lond. Math. Soc. **8**, 609 (1958).